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Mol. Cryst. and Liq. Cryst., 2001, Vol. 358, pp. 83-95
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Orientational Fluctuations and Pseudo-Casimir Force in Confined Nematic Liquid Crystals

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Our theoretical studies of fluctuations in the vicinity of phase and structural transitions and the fluctuation-induced force in two model confined nematic liquid crystals are briefly reviewed. We focus on the paranematic phase characterized by substrate-stabilized wetting layer and on nematic films frustrated by competing surface fields. Depending on temperature, anchoring strengths, and frustration, the fluctuation-induced force can be either short- or long-range, either attractive or repulsive, and it exhibits a pretransitional increase.

Keywords: confined liquid crystals; orientational fluctuations; fluctuation-induced force

INTRODUCTION

In the past decades, the use of microconfined liquid crystals in electro-optical applications has boosted the interest in the research of liquid-crystalline systems with high surface-to-volume ratio. In such systems, liquid-crystalline ordering is often assumed to be strongly distorted due to frustrating effects of the confining substrates, although in many cases this assumption has been found to be too naive.[1,2] However, a weak distortion of the ordering does not automatically imply that the structural force induced by the liquid crystal is weak too because in this regime the fluctuation-mediated pseudo-Casimir contribution to the force can be rather strong.[3] In the past few years the equilibrium structures in highly

frustrated geometries have been studied in detail,[4] whereas lately much efforts have concentrated on collective fluctuations and the fluctuation-induced force in these systems.[5–10]

In this paper we give a brief review of our recent studies of orientational fluctuations and structural forces in microconfined systems with various wetting and anchoring conditions.[7–10] In the next Section the model system and theoretical background are described. Then we address the wetting- and frustration-induced effects on fluctuations and pseudo-Casimir forces. In the last Section we interpret the results of a recent experimental study of the spinodal dewetting in terms of the fluctuation-induced force.[11]

THEORETICAL BACKGROUND

Microconfined liquid-crystalline systems are trapped in a variety of host materials characterized by curved, irregular, or even fractal internal geometry. Nevertheless, the confining geometries all share a common feature: the curved boundary and/or the antagonistic boundary conditions result in frustration which leads to a number of transitions between the equilibrium structures, the control variable being either temperature or the size of the system. Although the behavior of these systems can be very complex, the basic physics of the confinement-induced frustration can be modeled by the so-called hybrid nematic cell — a nematic liquid crystal film sandwiched between two parallel substrates inducing uniaxial nematic ordering but characterized by different wetting and anchoring properties.

To cover all aspects of the ordering, the nematic ordering is described by a tensorial order parameter. In the one-elatic-constant approximation, the associated Landau–de Gennes free energy density reads

$$f = \frac{L}{2}\left\{\xi_0^{-2}\left[\theta \operatorname{tr} \mathrm{Q}^2 - 2\sqrt{6}\operatorname{tr}\mathrm{Q}^3 + (\operatorname{tr}\mathrm{Q}^2)^2\right] + \nabla\mathrm{Q}\vdots\nabla\mathrm{Q}\right\}, \tag{1}$$

where L is the elastic constant and $\xi_0 = (27CL/B^2)^{1/2} \sim 10$ nm is the nematic correlation length at the bulk nematic-isotropic phase transition temperature T_{NI}, with B and C as standard Landau expansion coefficients.[12] θ is the reduced temperature such that $\theta = 0$ corresponds to the supercooling limit and $\theta = 1$ to T_{NI}, and the order parameter tensor Q is scaled by the degree of the nematic order at T_{NI}.[9] The surface part of the free energy density is described by the

tensorial analog of the Rapini-Papoular model,

$$f_{S_i} = \frac{L}{2\lambda_i}\operatorname{tr}(\mathrm{Q}-\mathrm{Q}_{S_i})^2\,\delta(z-z_i)\,. \tag{2}$$

Here λ_i's are the so-called extrapolation lengths which control the strength of the anchoring. Short extrapolation length, $\lambda_i \to 0$, corresponds to infinitely strong anchoring, and $\lambda_i \to \infty$ corresponds to infinitely weak anchoring. Q_{S_i} is the preferred value of the tensor order parameter on the i-th substrate; the substrates are located at $z_1 = 0$ or $z_2 = d$. In such a model, the surface interaction controls both the orientation of the nematic director and the degree of nematic order. Deep in the nematic phase, the tensorial Landau–de Gennes description can be replaced by the director picture and Frank elastic formalism consisting of splay, bend, and twist deformation of the director field.[13]

Within the mean-field approximation, the equilibrium configuration of the system is determined by the minimum of the free energy. Usually, the thermal fluctuations of the order parameter can be assumed small, and the free energy of fluctuations can be considered a correction to the mean-field free energy. In such a case, the fluctuations of liquid-crystalline order are described consistently by harmonic Hamiltonian of the form

$$H[b] = \frac{L}{2}\left\{\int\left[(\nabla b)^2 + \xi^{-2}b^2\right]\mathrm{d}V + \sum_{i=1,2}\lambda_i^{-1}\int b^2\mathrm{d}S_i\right\}, \tag{3}$$

where b stands for any of the five independent fluctuating degrees of freedom of the tensor order parameter (order parameter, biaxial, and director modes)[9] and ξ is a generalized correlation length characteristic of a particular type of fluctuations. In the case of uniform order parameter tensor the relevant temperature-dependent correlation lengths can be easily identified. In the nematic phase the order parameter fluctuations are characterized by

$$\xi_{N,o} = \xi_0\left[\frac{9}{4}\left(1+\sqrt{1-8\theta/9}\right)\sqrt{1-8\theta/9}\right]^{-1/2}. \tag{4}$$

The correlation length of biaxial fluctuations reads

$$\xi_{N,b} = \xi_0\left[\frac{27}{4}\left(1+\sqrt{1-8\theta/9}\right)\right]^{-1/2}, \tag{5}$$

whereas the correlation length of the director modes is infinite:

$$\xi_{N,d} \to \infty\,. \tag{6}$$

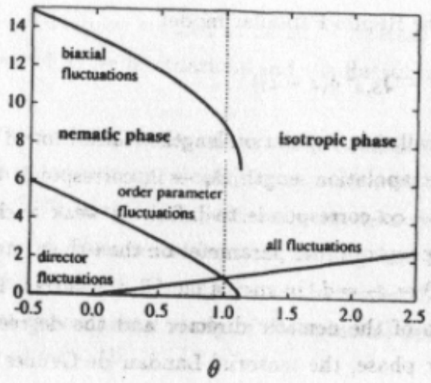

FIGURE 1 Correlation lengths of the three types of fluctuation modes vs. temperature. In the isotropic phase, all types of fluctuations are degenerate. In the nematic phase, director fluctuations correspond to the broken symmetry variable, and their correlation length is infinite.

The same Hamiltonian also describes fluctuations in the isotropic phase, where all five modes are degenerate and their correlation length is given by

$$\xi_I = \xi_0 \theta^{-1/2} . \tag{7}$$

The temperature variation of correlation lengths is plotted in Fig. 1.

Structural force

In planar geometry, the structural force between the substrates is defined by

$$\mathcal{F} = -\left(\frac{\partial F}{\partial d}\right)_{V,S} , \tag{8}$$

where F is the total free energy of the system in question and d is the separation between the substrates. As noted above, the total free energy includes the mean-field part and the free energy of fluctuations; the latter is given by

$$F_{CAS} = -k_B T \ln\left(\int \mathcal{D}b \exp(-H[b]/k_B T)\right) , \tag{9}$$

where k_B is the Boltzmann constant and T is the temperature.[14]

When discussing structural forces in confined liquid crystals one should bear in mind that there are two sources of the mean-field interaction. Elastic deformations of the director field cause a long-range force in the nematic phase, whereas variations of the degree of nematic order and biaxiality result in short-range force both in nematic and in isotropic phase. In both phases, due to the

restricted geometry the spectrum of fluctuations differs from the bulk spectrum, which results in additional fluctuation-induced force. Depending on the boundary conditions,[5,6] the fluctuation-induced interaction can be either repulsive or attractive, and its magnitude depends strongly on the surface interaction. In general, the sign of the pseudo-Casimir interaction is determined by the type of the boundary conditions, provided that the system is not subjected to electric or magnetic field. Using Eqn. (3) one finds $b(z = 0, d) = (\lambda/d)\, b'(z = 0, d)$, where $b' = \mathrm{d}b/\mathrm{d}z$. The fluctuation modes constrained by strong ($\lambda_{1,2} \ll d$) or weak ($\lambda_{1,2} \gg d$) anchoring at both substrates lead to an attractive force. In a mathematical language this corresponds to Dirichlet, $b(z = 0) = b(z = d) = 0$, or Neumann, $b'(z = 0) = b'(z = d) = 0$, boundary conditions at both substrates, respectively. In contrast the asymmetric situation with one surface enforcing a strong anchoring and the other a weak anchoring yields a repulsive force (mixed boundary conditions).[5,6]

In the following Sections the physics of fluctuations in the model system and the corresponding pseudo-Casimir force will be discussed briefly; the details of the calculation techniques have been described elsewhere.[7–10]

HETEROPHASE SYSTEM: NEMATIC WETTING IN ISOTROPIC PHASE

A simple example of a heterophase liquid-crystalline system is a paranematic film at temperatures slightly above the nematic-isotropic transition. The term paranematic phase is used for the state where surface or even electromagnetic fields induce partial nematic order, so that the liquid-crystalline ordering within a thin wetting layer is nematic rather than isotropic. In a semi-infinite sample the thickness of the wetting layer l diverges on approaching the nematic-isotropic phase transition as long as the surface coupling is strong enough (the so-called complete wetting regime).[15] Thus the paranematic system is in a heterophase state only very close to T_{NI} when $l \gg \xi_{N,o}$ ($\xi_{N,o}$ being the measure for the thickness of the interfacial area). The heterophase state becomes energetically unfavorable when l approaches $d/2$, which results in the transition to nematic state.

The collective dynamics of a heterophase system is characterized by two soft order parameter modes localized at the phase boundaries (Fig. 2). The modes

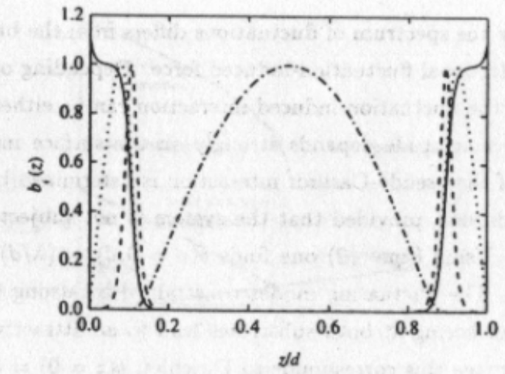

FIGURE 2 Mean-field degree of the nematic order (solid line) and lowest order parameter (dashed line), director (dotted line) and biaxial (dash-dotted line) modes in the paranematic phase just above the nematic-isotropic phase transition temperature. The soft order parameter mode is localized at the phase boundary, the lowest director mode is confined to the wetting layers, whereas the lowest biaxial mode is expelled from the wetting layers. $[d/\xi_0 = 100, \theta = 1.00001, \lambda = 0]$

correspond to fluctuations of thickness and position of the central isotropic part of the system; in a thick enough film ($d \ll \xi_{N,o}$), the two modes are practically degenerate. The lowest director fluctuation modes, which are confined to the wetting layers, also exhibit critical slowdown on approaching the clearing point. On the other hand, the biaxial modes are energetically cheaper in the isotropic phase and thus expelled from the wetting layers.

An insight into the nature of the dominant fluctuation modes in the paranematic phase will help us to understand the fluctuation-induced force in this system, which turns out to consist of two contributions: (i) the interaction between the substrates and the phase boundaries and (ii) the interaction between the two phase boundaries.[7]

Interaction between solid substrate and phase boundary

The interaction between the solid substrate and phase boundary consists of three contributions corresponding to three non-degenerated fluctuation modes. The fluctuations of the degree of order give rise to a short-range repulsion between the substrate and the phase boundary proportional to $\exp(-2l/\xi_{N,o})$, where l is the thickness of the wetting layer. The short range of the interaction is a consequence of finite correlations of the order parameter fluctuations in both

nematic and isotropic phase. The repulsion between the substrate and the phase boundary can be understood in terms of boundary conditions. The anchoring at the substrates is strong, whereas the maxima of the order fluctuation modes at the phase boundaries indicate that these modes experience effectively weak "anchoring" conditions. Because of the dissimilarity of the boundary conditions the interaction due to fluctuation of the degree of order is repulsive. A similar argument applies to biaxial fluctuations. Here weak "anchoring" condition at the phase boundary can be understood as a consequence of the fact that biaxial fluctuations are much more favorable in the isotropic phase than in the nematic phase. The resulting repulsive interaction is proportional to $\exp(-2l/\xi_{N,b})$ and thus much weaker than the order parameter modes' contribution because the correlation length of biaxial fluctuations is shorter than the correlation length of order parameter fluctuations. The main contribution to the interaction between the solid substrate and the phase boundary is induced by the director fluctuations, which are characterized by an infinite correlation length in the nematic phase. The leading term of this interaction reads[7]

$$-\frac{k_B T S \zeta(3)}{4\pi l^2}, \tag{10}$$

where ζ is the Riemann zeta function. This long-range interaction is attractive which can again be interpreted in terms of (dis)similarity of boundary conditions. In the isotropic phase, the director fluctuations are very "hard" compared to the ones in the nematic phase ($\xi_{N,d} \gg \xi_{I,d}$). Therefore the lowest normal modes are actually confined to the nematic surface layer.[9] The effective boundary condition at the phase boundary is very similar to strong anchoring at the solid substrate, and the force induced by director fluctuations is attractive.

Interaction between phase boundaries

The interaction between the two phase boundaries gives rise to an attractive fluctuation-induced force (identical boundary conditions) which is proportional to $\exp(-2(d-2l)/\xi_I)$. Except in the vicinity of the metastability limit of the paranematic phase, the distance between the substrate and the phase boundary is much smaller than the distance between the two boundaries, and the interaction between the phase boundaries is very weak. It should be also stressed that the range of the pseudo-Casimir interaction between the phase boundaries is half of the range of the mean-field interaction. Thus for $l \ll d$ the attractive mean-field contribution is dominant and proportional to $\exp(-(d-2l)/\xi_I)$. In very thin

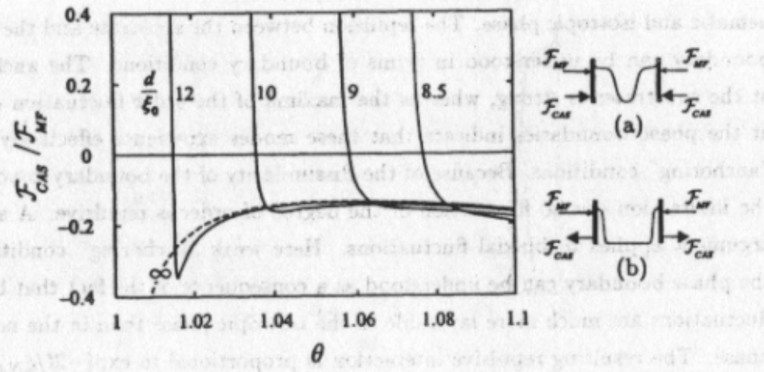

FIGURE 3 Fluctuation-induced force in the paranematic phase compared to the mean-field force as a function of temperature for $d/\xi_0 = 8.5$, 9, 10, and 12 (solid line, metastable state), whereas $d/\xi_0 \to \infty$ (dashed line; stable state). In general, the pseudo-Casimir force is controlled by fluctuations within the wetting layer (b) and thus repulsive. However, in the immediate vicinity of the metastability limit the force becomes dominated by the attraction between the phase boundaries (a).

cells (close to the metastability limit, $d/\xi_0 \sim 10$) the distance between the two phase boundaries becomes comparable to the thickness of the wetting layers and at certain point the paranematic state becomes unstable. In this range the attractive fluctuation-induced interaction between the phase boundaries becomes dominant.

Effective interaction between solid substrates

The effective pseudo-Casimir force between the two solid substrates is a superposition of the two contributions discussed above. In the range of stable paranematic phase ($\theta > 1$, $l \ll d$) the fluctuation-induced force between the two substrates is governed by the interaction between the solid substrate and the phase boundary, which is not directly measurable. In fact, it is mediated by the mean-field interaction which dominates the interaction between the two phase boundaries. Formally this enters via functional dependence of the wetting layer thickness l on the sample thickness d. Within the mean-field description, $\partial l/\partial d \approx -\text{const.} \times \exp(-d/\xi_I)$,[16] so that the leading term in the substrate-to-substrate fluctuation-induced force

$$\mathcal{F}_{CAS} \approx -\frac{k_B T S \zeta(3)}{2\pi l^3} \frac{\partial l}{\partial d} \propto \exp(-d/\xi_I) \tag{11}$$

is repulsive and short-range. Its range, ξ_I, is identical to the range of the mean-field force, whereas its magnitude is smaller but comparable to the magnitude of the mean-field attraction originating from the inhomogeneity of the order between the phase boundaries (Fig. 3). The two forces have the same range, so that the mean-field force is proportionally diminished by fluctuation contribution. The repulsive contribution of fluctuations can be perhaps most simply understood taking into account the fact that pseudo-Casimir attraction reduces the thickness of the wetting layers. The resulting increase of the distance between the two phase boundaries consequently reduces the effective interaction between the two substrates. In very thin films ($d/\xi_0 \sim 10$) close to the limit of the metastable paranematic region the situation is quite different because the pseudo-Casimir interaction between the phase boundaries becomes more important. Eventually, this interaction becomes dominant and at the metastability limit, the effective substrate-to-substrate interaction exhibits a attractive singularity (Fig. 3).[7]

ANCHORING-INDUCED FRUSTRATION: HYBRID NEMATIC CELL

In a hybrid nematic cell the confining substrates are characterized by mismatched easy axes; often one easy axis is homeotropic and the other is planar. Usually, dissimilar substrates differ in the anchoring strength as well. The competing surface interactions can result in several structures. In the vicinity of the clearing point, the equilibrium ordering in thin cells is biaxial and highly inhomogeneous.[10] On the other hand, deep in the nematic phase and for samples thicker than few $\xi_{N,o}$'s the degree of nematic order is almost uniform and uniaxial, whereas the director field can be either bent (thick cells) or uniform (thin cells). In films thinner than $d_c = |\lambda_P - \lambda_H|$ the uniform director structure is stable, whereas for $d > d_c$ the director field within the cell is distorted (λ_P and λ_H are the extrapolation lengths at the planar and the homeotropic substrate, respectively). In this review, we limit the analysis to films thick enough for the ordering to be consistently described by the director. We assume that the homeotropic anchoring is stronger than the planar anchoring so that the uniform structure is homeotropic.

In the following, we focus on the simplest ordered structure in the hybrid cell — the uniform structure. Its mean-field free energy consists of the energetic penalty for the violated boundary conditions at the planar substrate

$$F_{MF} = \frac{KS}{2} \lambda_P^{-1}, \tag{12}$$

which does not depend of the cell thickness and hence the mean-field force between the substrates is zero. In other words, in the uniform configuration the structural force is induced solely by fluctuations of the liquid-crystalline ordering.

The structural transition from uniform to distorted configuration is dominated by director fluctuations. Within the harmonic approximation Eqn. (3) the free energy of the two director modes can be determined by calculating the corresponding partition function [Eqn. (9)]. A detailed discussion of the methods of calculation of the partition function and the pseudo-Casimir force is beyond the scope of this paper. We shall only quote the analytical approximations and numerical results; a detailed analysis of the problem is discussed elsewhere.[8] To keep the analytical approximations as transparent as possible, we restrict our discussion to strong anchoring at the homeotropic substrate, i.e., $\lambda_H \to 0$ and $d_c \approx \lambda_P$. At small thicknesses, $d/d_c \ll 1$, the fluctuation-induced force is given by

$$\mathcal{F}_{\text{fluct}}(d/d_c \ll 1) \approx \frac{k_B T S}{2\pi}\left[\frac{3\zeta(3)}{8d^3} + \frac{\ln 2}{\lambda_P d^2}\right], \tag{13}$$

where ζ is the Riemann zeta function. The first term is the d^{-3} repulsion typical for mixed boundary conditions. The second term is a correction originating from a weak but finite destabilizing surface field at the planar substrate. Since this substrate promotes fluctuations, it enhances the force, i.e., it gives rise to an extra repulsion. In the case of strong anchoring the pseudo-Casimir force is repulsive at all thicknesses up to d_c. On increasing the distance, the d^{-3} repulsion [Eqn. (13)] gradually fades out, levels off in the vicinity of the structural transition to the bent structure and eventually diverges as $d/d_c \to 1$ [Eqn. (14) and Fig. 4]

$$\mathcal{F}_{\text{fluct}}(d/d_c \to 1) \approx \frac{3k_B T S}{2\pi\lambda_P^3} \ln\left(3(1 - d/d_c)\right)^{-1}. \tag{14}$$

In real hybrid systems with both λ_P and $\lambda_H > 0$, the fluctuation-induced force is attractive at small d/d_c's, then it becomes repulsive and reaches a local maximum before the pretransitional singularity. If the two anchoring strengths are not very different, the repulsive mid-range maximum is absent and the attractive regime extends almost right up to the structural transition. Although this behavior may seem surprising at first, it can be easily understood in terms of (dis)similarity of boundary conditions. At very small thicknesses ($d/d_c \ll 1$) both extrapolation lengths are larger than the thickness of the cell, which means that both surface interactions are effectively weak. This means that the fluctuations experience

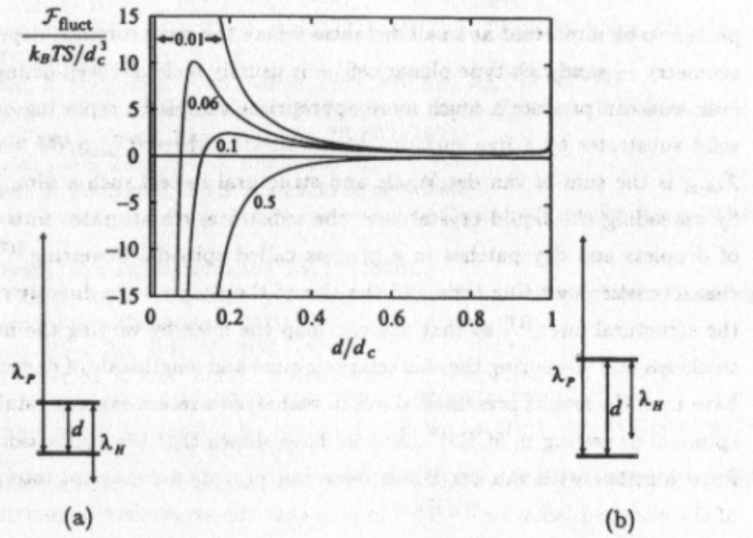

FIGURE 4 Fluctuation induced force in a hybrid cell as a function of the reduced thickness for various anchorings; $\lambda_H/\lambda_P = 0.01$, 0.06, 0.1, and 0.5. At small distances the fluctuation-induced force is attractive, $\lambda_P > \lambda_H > d$ [inset (a)], then it becomes repulsive, $\lambda_P > d > \lambda_H$ [inset (b)], and diverges at the transition ($d = d_c = \lambda_P - \lambda_H$).

effectively weak anchoring boundary conditions at both substrates, resulting in an attractive fluctuation-induced interaction. On increasing the distance, the fluctuation-induced force becomes repulsive when the homeotropic extrapolation length becomes smaller than the cell thickness: for $\lambda_H < d < \lambda_P$, the homeotropic anchoring is strong whereas the planar anchoring is weak (Fig. 4).

The results of the numerical analysis confirm the analytically predicted singularity of the fluctuation-induced force at the critical thickness. However, we stress that in the vicinity of the structural transition the anharmonic fluctuations may also play an important role. Nevertheless, the higher-order corrections are expected not to modify the divergent pretransitional behavior qualitatively.[8]

HAS THE PSEUDO-CASIMIR FORCE BEEN SEEN YET?

The main problem in observing the pseudo-Casimir force is its small magnitude compared to the mean-field force. Furthermore, the pseudo-Casimir force is ex-

pected to be important at small distances where the most common experimental geometry — sandwich-type planar cell — is usually no longer well-defined. However, one can produce a much more appropriate sample by replacing one of the solid substrates by a free surface. At thicknesses where $\partial \mathcal{F}_{\text{total}}/\partial d > 0$ (where $\mathcal{F}_{\text{total}}$ is the sum of van der Waals and structural forces) such a film, prepared by spreading the liquid crystal over the substrate, disintegrates into an array of droplets and dry patches in a process called spinodal dewetting.[17–19] The characteristic dewetting time and the size of the droplets are directly related to the structural force,[17] so that one can map the force by varying the initial film thickness and measuring the characteristic time and lengthscale of dewetting. We have used the results presented above to reanalyze a recent experimental study of spinodal dewetting in 5CB.[19], and we have shown that the fluctuation-induced force together with van der Waals force can provide a consistent interpretation of the observed behavior.[11] This implies that the experiment in question can be regarded as a first evidence of the pseudo-Casimir force in liquid crystals.

CONCLUSIONS

In this review, we summarized the results of our recent theoretical studies of fluctuations in two confined liquid-crystalline systems: the paranematic phase and the hybrid nematic cell. We showed that in these geometries, the behavior of fluctuations and the fluctuation-driven phenomena are governed by either stabilizing or destabilizing effects of the confinement. We analyzed the pseudo–Casimir force in both systems and we demonstrated that its confinement-induced features are very prominent, especially in the vicinity of phase and structural transitions. Some of the effects discussed may well have been observed indirectly in a recent study of spinodal dewetting of 5CB on a silicon substrate.

Acknowledgment

Support from Ministry of Science and Technology of Slovenia (Grant J1-0595-1554-98) and European Commission (TMR network No. ERB-FMRX-CT98-0209) is acknowledged.

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